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Title: Radioactive Material Dynamics @ NSLS-II

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Radioactive Material Dynamics @NSLS-II

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Bottom Line Up Front

- There is strong interest at NSLS-II for advancing DMMS capabilities for national security science, including accommodations for hazardous samples and/or classified work. A 3-phased approach is proposed:

Now	2 years	4 years
XPD Beamline	***HEX Beamline***	***MRE Beamline***
➢ Currently available ➢ 40-70keV (PDF 117keV) ➢ Radioactive including SNM samples ➢ XRD, SAXS, PDF	➢ Under construction (2 years) ➢ 30-150 keV mono (white beam up to 200 keV) ➢ ADXD, EDXD, SAXS, PCXI, CT, radiography	➢ MRE beamline (based on HEX) partnership (BES, INL/NE, NNSA) – proposed ➢ 7 wks/yr dedicated beamtime ➢ Dedicated experimental system and area for classified work ➢ Radioactive sample working area
	Investment: ~$20M	***Investment: $36M-$46M + operating***
Continue to develop and perform hazardous sample experiments – streamline safety/security process	*Design and construct specialized in-situ experimental system and sample environment for hazardous and/or classified samples*	*Partner in design, construction and operation of specialized beamline for materials science in extreme environments*

Note: all cost estimates are preliminary

"First Campaign"* Documents Describe DMMSC Motivations

- There are key types of materials vital to the nuclear deterrent:
  - Energetic Materials (high explosives), and understanding initiation in detail and impact of aging and defects.
  - Nuclear Materials (actinides), the mechanisms of aging, strength, compressibility, damage and failure.
  - Structural Materials (from foams and cushions to bottles, struts and supports), providing confidence to certify new processes for a life-time of use.
- We both care about understanding the performance of these materials over the lifecycle of the weapon, *and* we want to allow more flexible and reduced-cost options for manufacture of parts and production of systems.

* "(U) MaRIE First Campaigns," LA-CP-15-00501

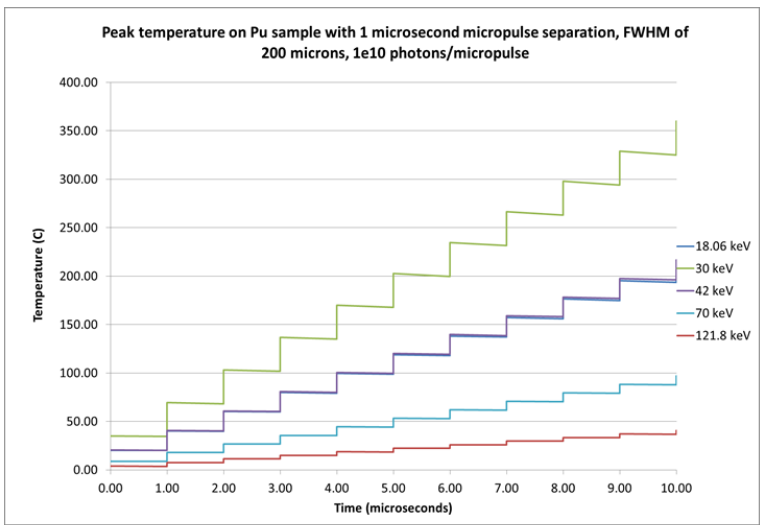
The Dynamic Mesoscale Materials Science Capability (DMMSC) addresses a national unmet scientific need for understanding material performance and production at the mesoscale.

The Radioactive Material Dynamics Endstation at NSLS-II MRE will Help Address Portions of the DMMSC Mission Need

RMD Enables: Classified work, SNM, 40-200 keV X-rays, flexible pulse structures for probing > μs dynamics with high-Z samples

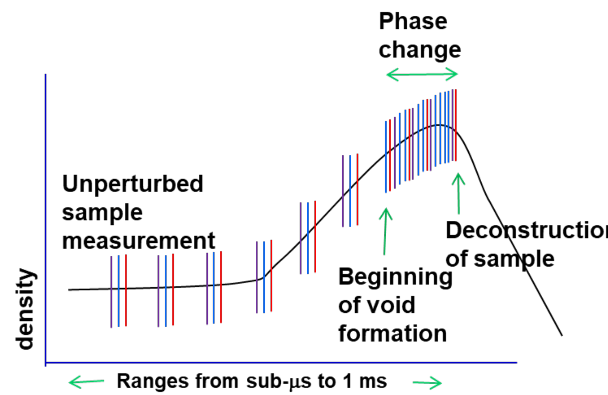
1) **Capacity to conduct classified experiments with SNM and relevant quantities of explosives**

2) **High energy X-rays (>42 keV) for measuring bulk high-Z properties by maximizing elastic scattering for diffraction and minimizing absorptive heating. (>70 keV recommended by the RMAB).**



These Scientific Functional Requirements were independently reviewed and formed part of the basis of the Requirements Management Advisory Board (RMAB) approval to proceed with an Analysis of Alternatives.

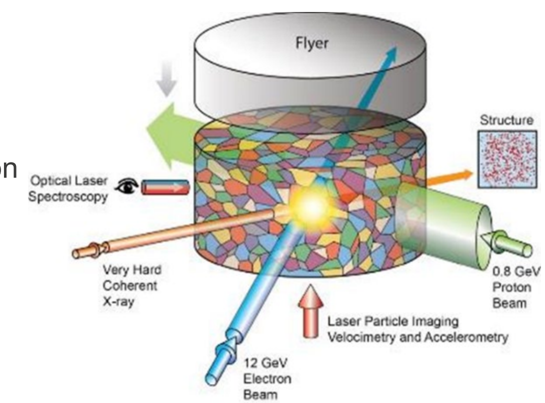
3) **A flexible pulse structure that can span electronic/ionic (sub-ps) to acoustic (ns) to shock transit (μs) to thermal (ms) event time scales for time-dependent control of the mesoscale.**



Pulse structure for shock observation depicted; welding and production require pulses to capture fast initial changes and slow cooling.

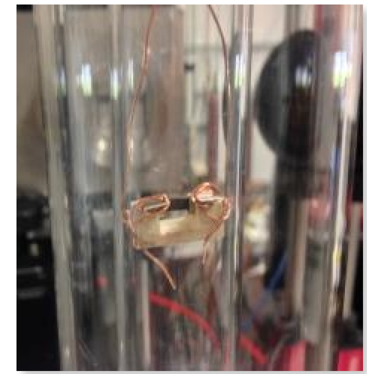
4) **Multiple probes to meet very different science needs**

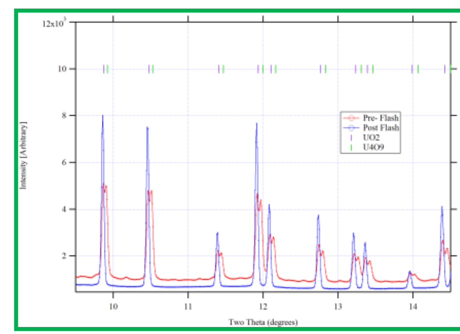
- Broad field-of-view: shock location, rarefaction waves, gross grain motion
- Narrow X-ray field-of-view: phase and grain plastic response
- X-rays, eRad, and pRad: differing areal densities
- Multi-view: single pulse ~3D information



DMMSC Mission Science Drivers

- Plutonium manufacturing science: thermo-mechanical processing, including microstructural evolution during deformation of Pu, alloys, and other materials for microstructurally-aware multi-phase strength models
- Pu phase data (changes induced by pressure, temperature), strength and damage testing through phases
- Uranium hydriding – initiation and evolution/propagation through material
- Fuel/cladding and actinide/other interactions (ex. species transport/thermal migration through materials)

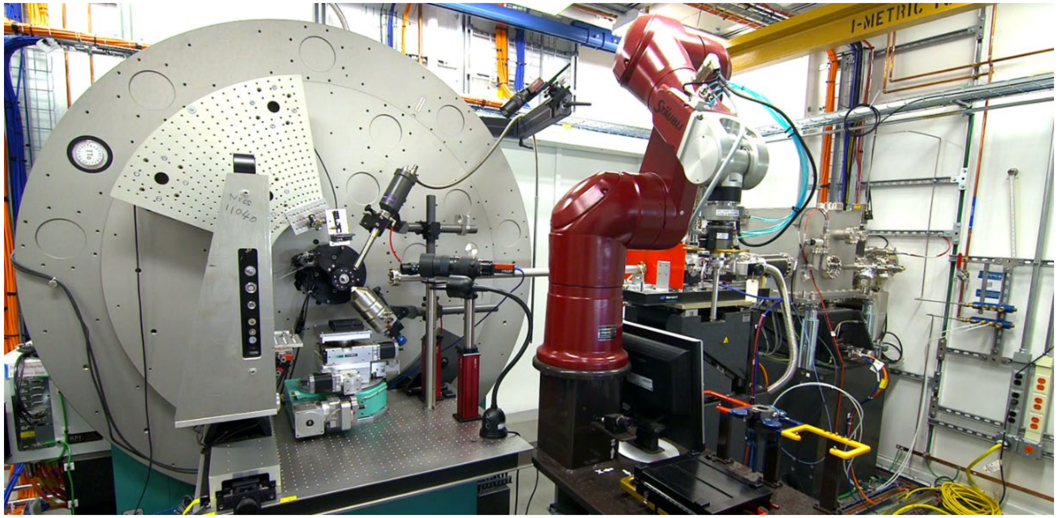




In Situ Synchrotron Characterization of Field Assisted Sintering of UO_2 at NSLS-II XPD show phase change with higher voltage and current

K. McClellan

XPD (X-ray Powder Diffraction)



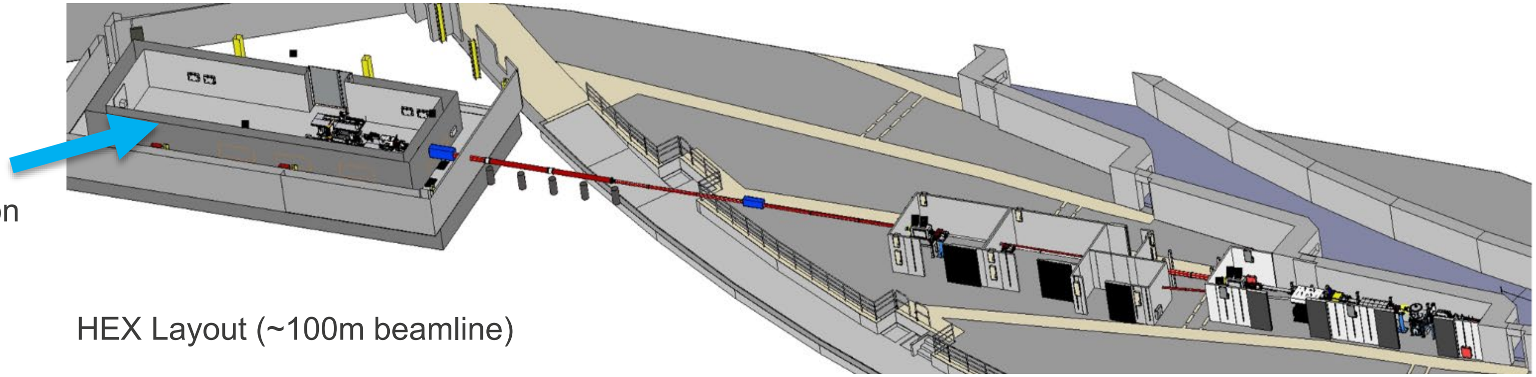
- Currently available
- Techniques: XRD, SAXS, PDF
- Parameters: 40-70 keV, beam size: 0.6 - 2 mm (h) x 0.2 – 2 mm (v), flux: ~6E13 ph/s
- Sample handling and environments for *in-situ* and *operando* conditions, robot for hazardous sample handling
- Previous and ongoing work with radioactive samples
- GU and “science commissioning” beamtime available
- Continue to develop and streamline process at NSLS-II for work with hazardous samples

The RMD *in-situ* Experimental System will Enable Collection of Quality Data on Hazardous Samples

- Self-contained experimental system/sample environment for radioactive material dynamics (RMD) that provides:
  - Ability to handle containment consistent with Pu and other radioactive material experiments
  - Multi-axis pressures of few kN (stress, torsion and shear forces)
  - Thermal loads from 10K – 3000K (note: most req. <2000K)
  - Vacuum, gas environment pressure/handling, composition monitoring
  - Sample manipulation and positioning
  - High-energy area detection (ex: Pilatus3 X CdTe)
  - Integrated controls systems (standalone and/or interfacing with beamline)

Timeline for design and construction: 2 years (commission at HEX)

HEX (High Energy Engineering X-ray Scattering)



HEX Layout (~100m beamline)

- Currently under construction (partner NYSERDA): completion of initial scope 2022
- Techniques: energy dispersive XRD (EDXD), mono angular dispersive XRD (ADXD), SAXS, PCXI, CT/radiography
- Parameters: 30-150 keV (1.3E11 ph/s/mm^2/0.1%BW @ 60keV), 20-200 keV white beam; beam sizes: 100 mm x 20 mm for imaging (resolution of 1 μm); 0.5 x 0.5 mm beam for diffraction, 10 μm micro-focus
- External building with large hutch that can accommodate NNSA Radioactive Material Dynamics (RMD) endstation (11 x 4.5 m^2 footprint available)
- GU beamtime

MRE (Materials in Radiation Environment)

Artist's rendition of MRE external building

- Propose to join a BNL-INL partnership (DOE-NE, establishing mission need) to develop and construct a beamline dedicated to the study of materials in radiation environments
- Significant design work performed, strong overlap of radioactive sample DMMSC and NE requirements
- External building enables increased security, higher level radiation area, rad work area
- Propose a dedicated hutch and experimental systems for classified work (of 4 potential hutches)
- Techniques: based strongly on HEX (reduced risk), potential for additional spectroscopy capabilities
- Parameters: 30-200 keV, μsec-msec time resolution, μm to mm beam sizes
- Transfer RMD endstation to MRE
- Dedicated beamtime (~7 weeks/yr) + GU time as needed

Timeline for design and construction: 4 years

A phased approach is planned for working with hazardous and/or secure samples and data

Beamline	Sample/Data	Facility	ESH Requirements
XPD (now)	Unclassified samples/data (VTR for secure analysis) Will establish LANL TACLANE	NSLS-II Security plan Current BL Staff, CI check	< 100 µCi/sample SNM levels < accountable < 0.5 g total ANM (for Pu-238 < 0.05g) 3 lvls containment (2 for solid) Ambient pressure and temp. Deviation requires extended review
XPD (w/ PPA)	As above, but data separated on local computers in PPA Investigate potential for occasional classified experiments	TPPA security plan, standalone PPA data room w/ electronic access control, barriers, sample data separation, surveillance protocols (det/cert not req'd), no sensitive country staff	As above, **increasing allowed SNM levels to ≥ accountable levels** with TPPA and existing NSLS-II Sample PPA
HEX	As above	As above	As above, **w/pressure and temperature as experimental parameters**
MRE	**Classified;** seek NSA TEMPEST waiver for Red/Black beamline/ accelerator separation; Multiple TACLANEs	**Classified area** per Intelligence Community Directive 705; NSA TEMPEST, DOE accredited. **Classified computing, VTC, sample handling and storage.** SSO, COMSEC, security staff. Cleared BL and support staff.	Accelerator Facility (not regulated under Nuclear Safety orders), separate safety basis including Accelerator Safety Envelope **Radiological inventory will be < Hazard Category-3 (DOE-STD-1027-92)**

Beam Specifications

At 100 keV

- 15-30 ps pulse length, 500 MHz operation
- 2E5 photons/pulse (10x more available in timing mode)

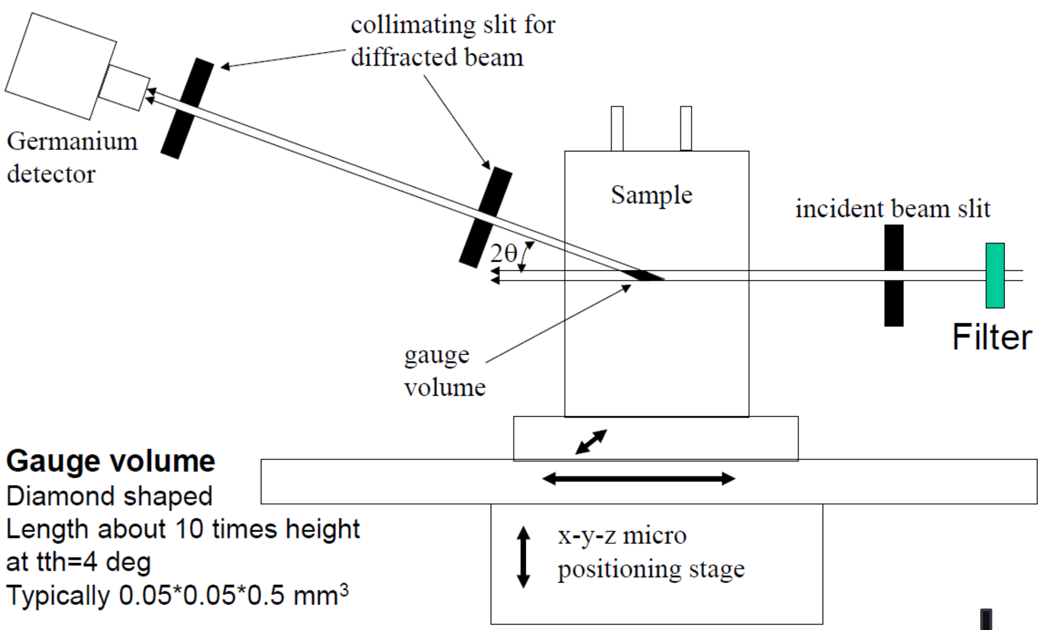
- Imaging beam: 8.0E10 ph/s/mm^2
  - Full field (100×20 mm^2) imaging with ≥1μm resolution using phase contrast CT
- Focused beam: 6.4E12 ph/s/mm^2

Example production science experiment: XRD observation at 100 keV of cooling from 1500 °C – 30 °C of a 1 mm thick UNb sample, with a cooling rate of ~20 °C/s

- For 1000 photons/pixel (on a Pilatus3 X CdTe detector at 1 m from sample), one would be able to take data at ~5 ms intervals

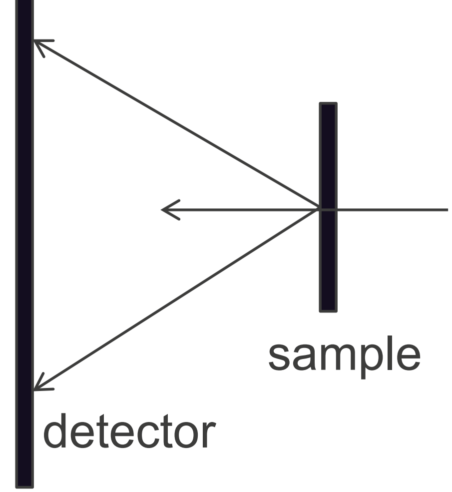
HEX & MRE will enable high energy x-ray imaging, XRD, and EDXD

Energy Dispersive X-ray Diffraction

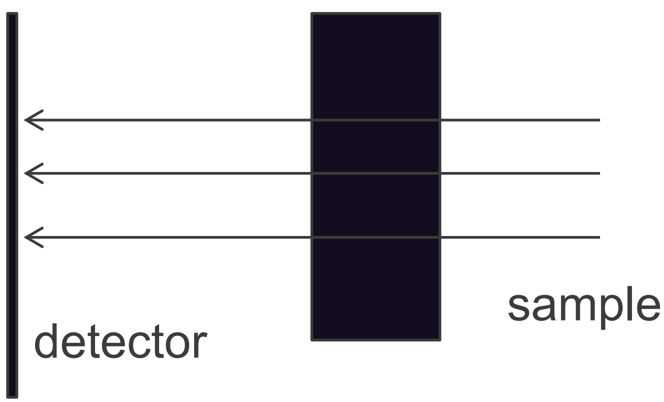


Gauge volume
Diamond shaped
Length about 10 times height at tth=4 deg
Typically 0.05*0.05*0.5 mm^3

Angle Dispersive X-ray Diffraction



Full field (100×20 mm^2) imaging with ≥1μm resolution using phase contrast CT



First experiments: *In-Situ* Uranium Hydriding to identify preferential sites for hydride nucleation and growth

Eloisa Zepeda-Alarcon, Samantha Lawrence, Don Brown (LANL)

Sample Environment

Depleted uranium cylindrical samples: 1mm diameter x ~10 mm tall.

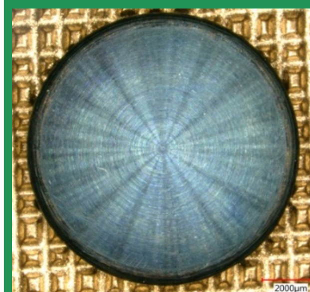
In-situ hydrogen gas dosing apparatus (provided by user). Pressure ~10-200 torr.

High temp capabilities up to ~500˚C.

A glove box capable of handling radiological material.

In-situ gas dosing apparatus at SMARTS beamline at LANSCE

Extended lifecycles (service and storage) for uranium components necessitate enhanced understanding of material corrosion and aging phenomena.



The current state-of-the-art techniques for studying uranium corrosion *via* hydride growth can not capture data at the mesoscale.

Synchrotrons can!

Required Techniques

Energy range: 70 keV – ~140 keV

Micron to sub micron spatial resolution.

Temporal resolution: 10^2 – 10^4 s/meas.

Micro-CT: hydride nucleation happens in the micron to sub micron length scales.

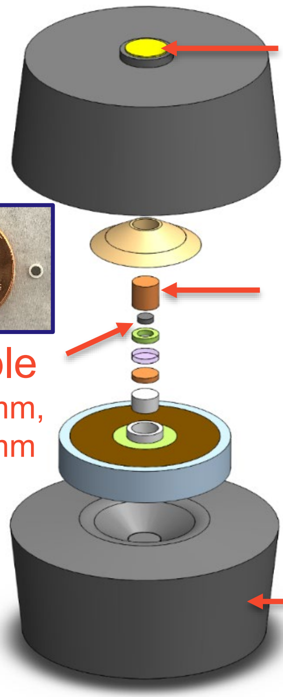
High energy x-ray scattering tomography: Map crystallographic phases and lattice strain evolution.

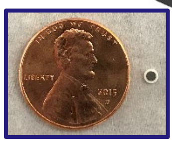
Far field high energy diffraction microscopy: Strain state of preferential sites

First experiments: Plutonium under pressure

Blake Sturtevant (LANL)

Large Volume High Pressure Experimental Platform





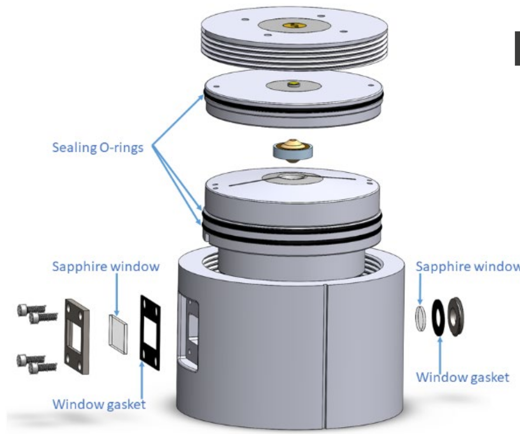
$LiNbO_3$
10° Y-rotated
50 MHz longitudinal
30 MHz shear

Al_2O_3
Buffer Rod

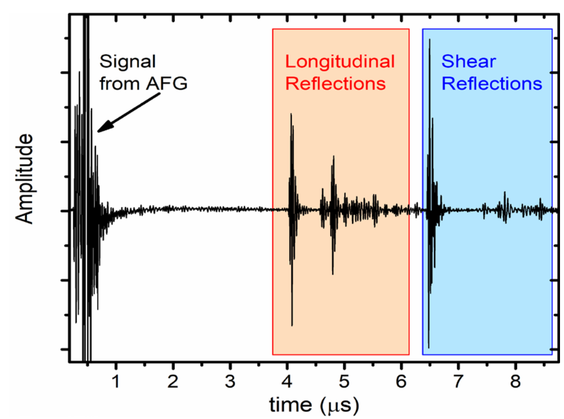
Sample
d=1.0 mm,
h=0.6 mm

Tungsten Carbide
High Pressure Anvil

An ultrasound transducer excites sound waves through the anvils to determine elastic moduli as a function of pressure and temperature



3x Containment Vessel

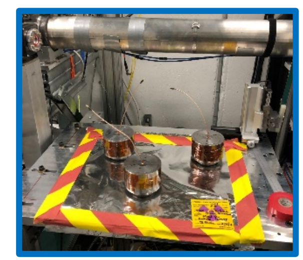


Experimental Techniques

Ultrasound

Energy Dispersive X-ray Diffraction

Radiography

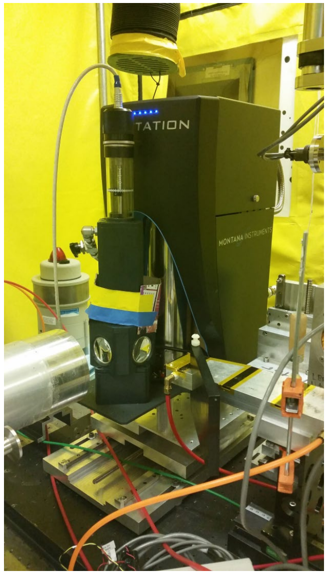


Successful feasibility demonstration of *in-situ* experiments at APS HPCAT

Requires larger Pu sample sizes to answer scientific questions

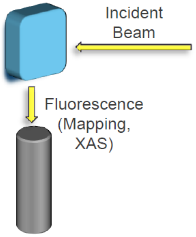
First experiments: Understanding fundamentals of radiation damage in Pu will inform models of aging phenomena for stockpile stewardship

Dan Olive (LANL)



Experimental Technique: Extended X-ray Absorption Fine Structure (EXAFS): Element-specific local structure information

- Study structural changes occurring in the first few years after Pu production
- Study effect of impurities on newly cast materials (production science)
  - Accelerated damage production using cold storage
  - Self-annealing mechanisms in δ-Pu alloys from 5°K-RT
  - Induced annealing at higher temperatures



Need for larger sample masses to inform and validate models

MRE can enable Pu K-edge EXAFS on relevant sample sizes

First Experiments: Qualification and Certification of Pu Manufacturing – the path to "Born-qualified"

Jason Jeffries (LLNL)

- We will need to design **new weapons systems** with new constraints
- **How to qualify** these? Process evaluation takes 5-10 years per process (plus certification)
- To become flexible/agile, **develop a "Born-qualified" approach**
- Multiple Tools: Casting, rolling, AM – choose the right one for the task
- **Model a process for desired property/performance** to shorten dev. time
- ***Need to show that these models are valid***
- Experiments (Diffraction, Imaging):
  - Small casting apparatus to study *in-situ* solidification, grain growth, chemical homogenization/solutionizing using relevant surface/volume ratios
  - Small deformation rigs to understand phase changes, stress concentrations and microstructure during rolling
  - *In-situ* weld diagnostics (local stress, strain, structure, grain size) for model development

RMD @ MRE will enable classified *in-situ* studies of Pu manufacturing processes

Radioactive Materials Dynamics @ NSLS-II

- Proposal to develop radioactive material dynamics capabilities at NSLS-II leverages strong overlap and investment from NE and BES to meet DMMSC mission needs.

Now	***2 years***	***4 years***
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Note: all cost estimates are preliminary

RMD @ NSLS-II helps pave the path to a DMMSC

- Developing expertise in techniques and experimental methods for DMMSC science
- Improving capabilities for models to become microstructurally aware
- Workforce training and development
- Help refine DMMSC scientific requirements
- Complementary to DMMSC XFEL science (EDXD/EXAFS)